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Mathematical modelling of the steam stripping of aroma from roast and ground coffee

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Abstract

Instant coffee manufacture involves the aqueous extraction of soluble coffee components followed by drying to form a soluble powder. A challenge that arises from the process is the loss of volatile aroma compounds during evaporative drying. One method of retaining aroma is to first steam strip the volatiles from the coffee and add them back to a concentrated coffee solution just before the final drying stage. Understanding the impact of process conditions on the aroma content of the stripped solution offers engineers the ability to target desirable compounds and maximise their yield. This paper presents a multiscale model for aroma extraction that describes: (i) release from the matrix, (ii) diffusion through the coffee grain, (iii) transfer into water and steam, and (iv) advection through the stripping column. Results reveal how aroma physiochemistry determines the limiting kinetics at industrial extraction conditions. The interaction with the soluble matrix can also inhibit extraction, but this effect varies for the different aromas studied.

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Keywords: Coffee; aroma; flavour; steam stripping; advection-diffusion model

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1. Introduction

Coffee is the most popular prepared beverage, with global consumption of over 9.5 million tonnes of green coffee beans [1,2]. Instant coffee dominates the UK coffee market with a share of 79.4%, equivalent to over 47000 tonnes [3].

The instant coffee process begins with the roasting of green beans to develop flavour compounds [4]. Roasted coffee beans undergo particle size reduction and then multiple stages of high temperature aqueous extraction of the soluble components. This extract is dried by evaporation and spray or freeze drying to form a soluble powder.

To reduce aroma degradation and loss caused by thermal processing, an aroma stream is extracted immediately after grinding, which can be added to the concentrated coffee extract before the final drying stage. To produce a concentrated aroma extract, techniques [5-7] that are based on interaction between ground coffee, water and/or steam are described in the patent literature. These aroma management steps determine the final product aroma content, therefore the engineer must understand the mechanisms by which aroma transfers from coffee grounds into aqueous and gaseous media, so that processes can target the desired aroma content.

Aroma exists principally within the coffee oil [4]. There will be some easily accessible aromas on the surface of the grain, which will dissolve in the water and partition into the headspace. From there the aromas will be carried by the advection of steam. As the surface is depleted and as water fills the pores, aroma will continue to dissolve and diffuse to the surface.

A study of the kinetics of aqueous extraction, for domestic coffee brewers, showed how hydrophilic aroma compounds extract significantly faster than lipophilic ones [8]. In another study, when stripping aroma with gas from a dry coffee bed, data could be fitted to the analytical solution of a Fickian diffusion model [9]. However, upon wetting the coffee, the behaviour of some compounds could not be described using the diffusion model. The addition of the aqueous phase seems to introduce new mechanisms – and thus complexity – to extraction, like the interaction with other soluble components i.e. binding of different food components directly to aroma compounds or to free water [10]. Coffee extraction has been modelled too by describing transfer between inert coffee solid matter, coffee particle (intragranular) pores and coffee bed (intergranular) pores. The work in Ref. [11] defines diffusion lengths between each phase, and uses fitted transfer coefficients to describe these processes.

There is, to date, no published model of aroma extraction derived from first principles that solves the particle scale diffusion equation and bed scale advection simultaneously. The aim of this work is to provide such mathematical model, and, from it, reveal the different rate-limiting extraction mechanisms for different compounds and predict how features of the process within (or without) the control of the engineer can impact the chemical and sensory profile of the resulting distillate. This will provide a useful tool to design processes that optimise aroma yield, concentration and desired sensory attributes.

Nomenclature

A_{bed}	cross-sectional area of the bed [m^2]
a_{12}	surface area of 1 per unit volume of 2 [m^{-1}]
c	concentration [mol m^{-3}]
D_{eff}	effective diffusivity [m^2 s^{-1}]
d	diameter [m]
H	Henry's law constant [Pa m^3 mol^{-1}]
$K_{o/w}$	octanol-water partition coefficient
k	mass transfer coefficient [m s^{-1}]
Q	volumetric flowrate [m^3 s^{-1}]
R	gas constant [J mol^{-1} K^{-1}]
r	radial position [m]
T_b	bed temperature [K]
t	time [s]
Z	bed height [m]
z	bed position [m]

Subscripts

b	bed
do	oil phase
g	gas phase
PP	polyphenol
p	particle (free phase)
por	mesopore
w	water phase

2. Extraction process

This work focuses on a soluble coffee manufacture process like that described in [12]. Roasted and ground coffee beans (<1.8 mm diameter), are bed-packed in a cylindrical column (Fig 1a). A defined quantity of water is first added, which is assumed to be perfectly distributed through the column and is sufficient to entirely fill the pores in the coffee particles, wetting up to 70% moisture content. Saturated steam enters via the column base for up to 40 minutes, and a vacuum (as low as 0.1 bar) is applied at the top, causing plug flow advection that carries aroma up the column. In each column section, the bed consists of loose-packed particles, with free water existing as a surrounding uniform, stagnant film (Fig 1b). Henry's law constants describe the partition into the headspace and there is convective transport into the steam across the surface area of the particles (of order10 m2 kg-1). A diffusive flux determines the transfer from particle surface to free water in the bed. Each particle is a porous matrix, with porosity consisting of large spherical pores connected by a nanoporous network through the cell wall (Fig. 1c). Mass transport within the particle (c.1 mm radius) combines free diffusion in the mesopores and hindered diffusion in the cell wall (c.10 μm thickness). The aroma is dissolved in a thin layer of oil (c.1 μm thickness) which coats each large spherical pore (Fig. 1d), and partitions and diffuses across the mesopore radius into the free phase according to the octanol-water partition coefficient.

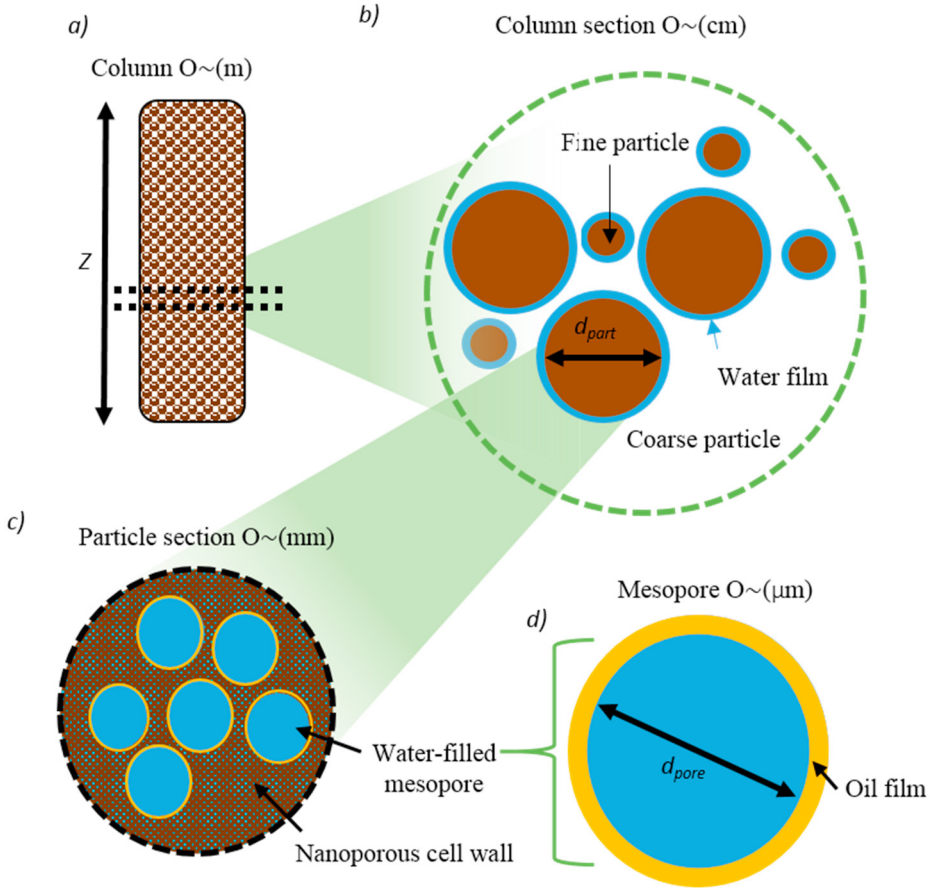

Fig. 1. Schematic diagrams of different scales in the system with key geometries (a) whole column, with labelled height (Z); (b) column section, with labelled bed pore size ($d_{b,pore}$) and particle diameter (d_{part}); (c) particle section with mesopores distributed in a nanoporous matrix all filled with water; (d) mesopore with a labelled diameter (d_{pore}) and oil thickness (δ).

3. Extraction model formulation

A multiscale model, coupling transport phenomena and aroma kinetics, has been derived from first-principles to describe the extraction process detailed in Section 2. The model formulation describes aroma mass transfer in the packed bed, as well as mass transfer of aroma and soluble matrix components at particle scale. The main assumption used for each scale are detailed next.

3.1 Assumptions

For the bed scale:

- One particle from each size class is taken as being representative of the column section.
- Column is perfectly insulated.
- No radial variation in flow, temperature or water content.
- The dominant mechanisms of aroma mass transfer are diffusion (intra-particle liquid-liquid and particle surface liquid-gas) and advection in the gas phase.
- All material flowing out of the top of the column is condensed.
- The mobile phase is an ideal gas.

For the particle scale:

- Particles are spheres (so radial symmetry applies) of homogeneous porosity.
- Particles are bimodally distributed - the diffusion equation is solved for each class of coarse and fine particles.
- Porosity consists of spherical mesopores connected by a nanoporous network which is saturated with water.
- No swelling effects.
- Aroma initially exists dissolved in an oil layer uniformly spread within the mesopores, and dissolved aroma partitions according to the octanol-water partition coefficient.
- Aroma extraction is not limited by solubility.
- Physical properties (density, thermal conductivity etc.) are weighted averages of coffee and water properties.
- Free water in the bed exists as a homogeneous stagnant film coating the particles.

3.2 Bed scale aroma mass transfer

The concentration of each individual aroma is modelled independently. Initially there is no aroma in the water or gas phase and transfer to the gas phase does not begin until the steam contacts that column element. Equation (1) describes the concentration in the gas as the sum of advection and source terms; dispersion is considered negligible:

$$\frac{\partial c_g(z,t)}{\partial t} = -\frac{Q(z,t)}{A_{bed}\varepsilon_b}\frac{\partial c_g(z,t)}{\partial z} + k_g a_{bp}\left(c_w(z,t)\frac{H(T_b)}{RT_b(z,t)} - c_g(z,t)\right) \tag{1}$$

At the column inlet there is no aroma in the gas phase, while the outlet is an open boundary (the material flowing out is dependent upon the upstream conditions).

The concentration in the stationary free water in the bed is described in equation (2) by source and loss terms from/to the particle surface and gas respectively.

$$\frac{\partial c_w(z,t)}{\partial t} = \left(k_w a_w(z,t)\left[c_{p,R}(z,t) - c_w(z,t)\right] - k_g a_w(z,t)\left[c_w(z,t)\frac{H(T_b)}{RT_b(z,t)} - c_g(z,t)\right]\right) \tag{2}$$

The mass transfer coefficient k_g has been obtained from [13] for Re<1000. For the stagnant water layer, with Re~ 0 and Sh~ 2, the mass transfer coefficient k_w was calculated from [14].

3.3 Particle scale

3.3.1 Aroma mass transfer

Two particle-scale concentrations are modelled: (i) dissolved aroma in the oil layer of the mesopores and (ii) free aroma that diffuses through the particle in the water-filled mesopores. The release of dissolved aroma is described as:

$$\frac{\partial c_{do}(r,t)}{\partial t} = -k_{por} a_{por,do} \left(\frac{c_{do}(r,t)}{K_{o/w}} - c_p(r,t) \right) \tag{3}$$

where the mass transfer coefficient is calculated assuming free diffusion across stagnant water. The diffusion of free aroma is described using Fick's Second law with a source term from the dissolved aroma being released:

$$\frac{\partial c_p(r,t)}{\partial t} = D_{eff}(r,t) \left(\frac{2}{r} \frac{\partial c_p(r,t)}{\partial r} + \frac{\partial^2 c_p(r,t)}{\partial r^2} \right) + k_{por} a_{por,p} \left(\frac{c_{do}(r,t)}{K_{o/w}} - c_p(r,t) \right) \tag{4}$$

As there are two phases through which the mobile aroma diffuses – an unhindered mesopore region and a tortuous nanoporous cell wall – D_{eff} is calculated by using the Maxwell-Garnett homogenization model [15]. Values for the free diffusion coefficient were obtained from literature [16] for several compounds, and a correlation with molecular weight was made to estimate others. The diffusion coefficient for loosely packed beds is a function of particle porosity as defined in [17], and is a function of temperature and viscosity according to the Stokes-Einstein equation.

Initially, there is uniform aroma distribution in the oil layer within the mesopores, and no free aroma. Symmetry is assumed in the centre of the particles and a flux boundary condition is imposed on the boundary between the particle mesopores and surface water.

3.3.2 Soluble matrix as a sink for aroma

An irreversible reaction mechanism that is first order in each species, and with a 1:1 molar ratio of the reactants, is used to model the interactions occurring between aroma and the soluble matrix [18]:

$$c_p c_{p,PP} \xrightarrow{k_{on}} c_{p,comp} \tag{5}$$

For compounds undergoing this binding, an additional reaction term, $r = -k_{on} c_p c_{p,PP}$, is added to the diffusion and source terms of equation (4). The same reactions will occur in the free water phase outside of the particles. The polyphenol content, partition coefficient and diffusivity have been taken or estimated from literature [17,19].

4. Results and Discussion

4.1. Model validation

The model formed by equations (1)-(4) was solved using the Finite Difference method. A FTCS (Forward-Time Central-Space) scheme was implemented in MATLAB using N = 50 and 5 coarse and fine particle nodes respectively and N = 25, and results were validated using literature data [9]. Figures 2(a) and 2(b) compare model results with the published experimental data for small-scale aroma gas stripping of coffee, showing the model's suitability to describe 3-phase diffusion-limited and reaction-limited aroma extraction.

In addition, model results for industrial scale extraction were validated using the analytical solution for Fickian diffusion processes [15] in a XD geometry with a spatial mesh consisting of N = 100 and 10 coarse and fine particle nodes respectively and N = 100 column nodes (computing time ~ 20 min), which guaranteed a mesh independent solution.

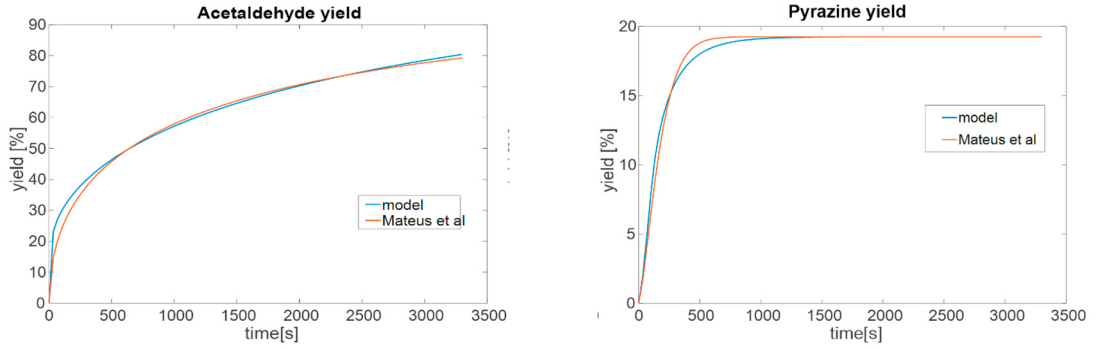

Fig. 2: Comparison between model and literature data [9] for two key aroma compounds displaying (a) acetaldehyde yield for diffusion-limited extraction and (b) reaction-limited kinetics of pyridine with binding rate constant of 8.12x10-4.

4.2. Steaming time

A range of process parameters may be employed to produce an aromatized distillate according to [12]. Using the model, we explore how variation in the parameters may cause significant changes in the distillate aroma profile. A simple process variable to control is the time taken to complete the steam strip. Several aromas with different properties (Table 1) were taken to study the effect of steaming time.

The concentration results of simulating a 20 min and 40 min steam strip process are shown in Fig 3. By doubling the length of time spent steaming the coffee, the model predicts a dilution in many aromas to between 50-75% of the concentration obtained after 20 min.

This suggests the extraction rate is slowing as the aromas become stripped from the particle surface and become internally-diffusion limited. For pyridine, which is consumed by binding reactions with the soluble coffee matrix, the concentration is <20% of the value after 20 min. The very apolar compound (β-Damascenone) after 40 min is only slightly more dilute than at 20 min, and a compound with a very low Henry's volatility partition coefficient (furaneol) is over 30% more concentrated at the longer steaming time compared to the shorter time.

4.3. Types of extraction kinetics

Simulation results suggest that the non-binding aroma compounds exist in three groups. Each group's extraction is limited by i) partition into the aqueous phase, ii) internal diffusion or iii) partition into the steam phase. An example of each characteristic extraction kinetic is shown in Fig. 4.

Table 1: Aromas under investigation, with their key sensory and physical properties

Aroma compound	Sensory note [20]	Octanol-water partition coefficient [21]	Henry's law volatility constant at 298 K [Pa mol^{-1} m^{3}] [21]
Beta-damascenone	Fruit, sweet, honey	4.2	22.8
2-fururylthiol	Coffee, roasted	3.4	5.34
Guaiacol	Earthy, burnt, smoky	1.3	0.259
Furaneol	Sweet, caramel	0.82	0.000536
Pyridine	Fish, Burnt	0.65	0.279
Acetic acid	Vinegar, sour	-0.17	0.0250

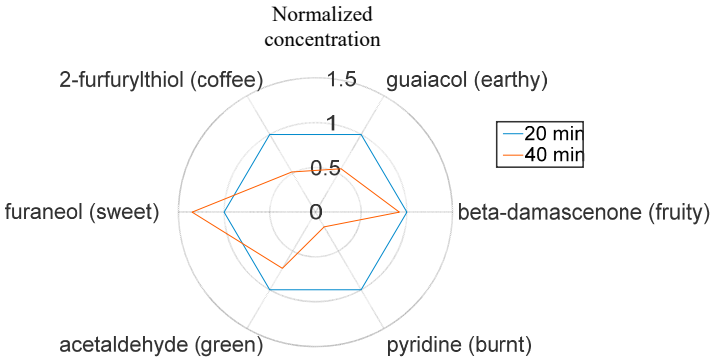

Fig. 3: Concentration (normalized to values after 20 min of steam stripping) of some key aromas in the distillate when simulating 20 min and 40 min steam stripping.

Type 1 - Low solubility. For very apolar compounds the aromas remain partitioned strongly in the mesopore oil layer because their extraction is limited by the low solubility in the aqueous phase. They require strong internal concentration gradients between pore water and oil to enhance the mass transfer rate. Over timescales of minutes, only mesopore water within the surface of the particles are depleted and set up these large concentration gradients. As a result, extraction is strictly limited by surface area and distillate concentration slowly falls with time.

Type 2 – Internal diffusion controlled. Most compounds fall into the second category. After an initial peak in concentration in both water and steam phases as extraction from near the particle surface takes place, the aroma is quickly stripped out and concentration in the gas falls. Extraction is subsequently determined by the particle features that impact internal diffusion.

Type 3 – Low volatility. Some very polar compounds have low Henry's law constants, and stay strongly partitioned in the water phase. They quickly reach saturation concentration in the steam and their extraction is limited by how much steam can be contacted with the water in the time available. As a result, their concentration increases slowly as material accumulates in the water (increasing the saturation concentration in the steam). Over the timescale of practical steam stripping, this is the observed effect, however, eventually diffusion limitations would lower the concentration.

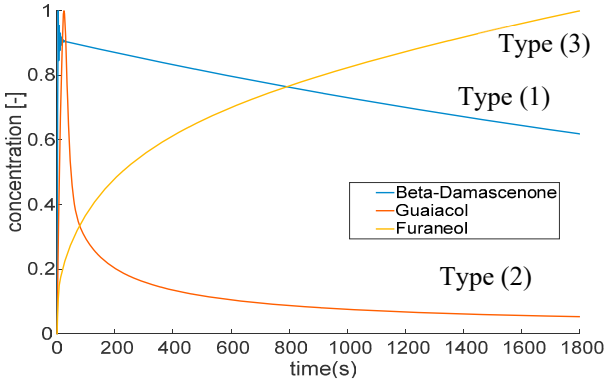

Fig. 4: Normalized concentration of aromas in the exiting steam demonstrating the limiting factors in extraction (time shown is after the column reaches saturation temperature).

4 Conclusions and Future work

This work presented a first principles-based model for aroma extraction during soluble coffee manufacture. This model spans scales from particle to column extraction, describing aroma: (i) release from the matrix, (ii) diffusion through the coffee grain, (iii) transfer into water and steam, and (iv) advection through the stripping column. Published data for extraction kinetics of different aroma compounds have been used to validate the model. Besides extraction

kinetics, the effect of different steaming times on aroma extraction has been also investigated. Results show that doubling the time the coffee is steamed leads to significant aroma dilution - between 50-75% of the concentration for a wide range of compounds. The model also allows the classification of aroma compounds according to their partitioning and binding behaviour, revealing three different limiting extraction kinetics, i.e.: (i) partition into the aqueous phase, (ii) internal diffusion and (iii) partition into the steam phase. Overall, these findings set the basis for the design of optimised extraction processes that target specific aroma profiles.

Further model development is aimed at including steam stripping of dry and partially wet coffee, which has the potential to lead to a more versatile and reliable predictive model for use in process design oriented to chemical and sensory outcomes. This model extension will be done by describing absorption of condensing steam, which enhances the extraction of certain compounds. Pilot plant experiments will be designed to validate the suitability of this model extension and the trends predicted in aroma extraction.

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